

On correlation approach to scattering in the decoherence timescale

Towards the theoretical interpretation of NCS and ECS experimental findings

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Abstract

We provide a "first principles" description of scattering from open quantum systems subject to a Lindblad-type dynamics. In particular we consider the case that the duration of the scattering process is of similar order as the decoherence time of the scatterer. Under rather general conditions, the derivations lead to the the following new result: The irreversible time-evolution may cause a reduction of the system's transition rate being effectuated by scattering. This is tantamount to a shortfall of scattering intensity. The possible connection with striking experimental results of neutron and electron Compton scattering from protons in condensed matter is mentioned.

Keywords: irreversible dynamics, entanglement, decoherence, neutron Compton scattering, electron-proton Compton scattering

1 Introduction

The counter-intuitive phenomenon of entanglement [1] between two or more quantum systems has emerged as the most emblematic feature of quantum mechanics. Experiments investigating entanglement, however, are mainly focused on collections of few simple (two- or three-level) quantum systems thoroughly isolated from their environment (e.g., atoms in high- Q cavities and optical lattices). These experimental conditions are necessary due to the decoherence of entangled states. In short, decoherence refers to the suppression of quantum superpositions caused by the environment. By contrast, entanglement in condensed and/or molecular matter at ambient conditions is usually assumed to be experimentally inaccessible. However, two new scattering techniques operating in the sub-femtosecond time scale provided results indicating that short-lived entangled states may be measurable in condensed matter even at room temperature [5, 6].

In this paper we provide a first-principles treatment of scattering from "small" open quantum systems in condense-matter environments, in the "time window" of decoherence of the scattering system. That is, the focus is in "fast" scattering processes with

a duration (usually denoted scattering time, τ_{sc}) of the order to the scatterer's decoherence time, τ_{dec} . This may be considered to represent an "extension" of standard scattering theory — as applied e.g. to neutron physics [2, 3] or electron scattering [4] — in which the concepts of entanglement and decoherence play essentially no role. The first part of the derivations are analogous to the standard (often denoted) "van Hove formalism" [2]; see also the textbook [3]. Then a reduced open quantum system, i.e. a micro- or mesoscopic system characterized by a set of preferred coordinates, is introduced. This corresponds to the "small" physical system that scatters a neutron (electron, etc.) with a sufficiently large momentum transfer. Its dynamics is described by a simple Lindblad-type master equation [7, 8] (which, for the sake of simplicity, contains only one Lindblad operator, X), thus including explicitly the effect of decoherence into the formalism.

The striking result of the derivation may be summarized as follows: The irreversible time-evolution (owing to the Lindblad operator X) may cause a reduction of the transition rate of the system (from its initial to its final state). In "experimental" terms, this is tantamount to an effective reduction of the system's cross-section density and thus a shortfall of scattering intensity.

2 Scattering in brief

We assume an N-body Hamiltonian $H_{total} = H_0 + V$ with an interaction of the form

$$V(\mathbf{r}) = \lambda n(\mathbf{r}) \quad (1)$$

where $n(\mathbf{r})$ is the particle density operator and λ is the rest of the interaction (contact potential). For example, in the case of neutron scattering from a system consisting of N particles with the same scattering length b one may put

$$n(\mathbf{r}) = \frac{1}{V} \sum_j \delta(\mathbf{r} - \mathbf{R}_j) \quad (2)$$

where V is the volume, \mathbf{R}_j is the spatial position of the j -th particle, and

$$\lambda = \frac{2\pi\hbar^2}{m} b, \quad (3)$$

m being the neutron mass. (For further details about scattering from "bound" and "free" particles, see the textbook [3].)

In the *interaction* picture, the Schrödinger equation is now (putting for simplicity $\hbar = 1$)

$$i\partial_t \Psi = \lambda n(\mathbf{r}, t) \Psi \quad (4)$$

with the perturbative solution

$$\Psi(t) = \Psi(0) - i\lambda \int_0^t n(\mathbf{r}, t') dt' \Psi(0). \quad (5)$$

We write the transition probability $W(t)$ between initial states ψ_i (with probability P_i that the scattering system is in the state ψ_i) and final states ψ_f of the scattering system to be given by

$$W(t) = \sum_{i,f} \left| \langle \psi_f | \lambda \int_0^t n(\mathbf{r}, t') dt' | \psi_i \rangle \right|^2 P_i. \quad (6)$$

It should be noted that ψ_i and ψ_f are eigenstates of the unperturbed N-body Hamiltonian H_0 [3, 2]. This allows us to write the transition probability in the form

$$W(t) = \lambda^2 \int_0^t dt' \int_0^t dt'' \sum_f \langle \psi_f | n(\mathbf{r}, t') \rho n(\mathbf{r}, t'') | \psi_f \rangle, \quad (7)$$

where

$$\rho = \sum_i |\psi_i\rangle P_i \langle \psi_i|, \quad (8)$$

by noting that $n^\dagger(\mathbf{r}, t) = n(\mathbf{r}, t)$, since \mathbf{R}_j and \mathbf{r} are Hermitian operators.

In an actual scattering experiment from condensed matter, we do not measure the cross-section for a process in which the scattering system goes from a specific initial state ψ_i to another state ψ_f , both being unobserved states of the many-body system. Therefore, one takes an appropriate average over all these states [3, 2], as done in Eq. (6).

Furthermore, the initial (\mathbf{k}_0) and final (\mathbf{k}_1) momenta of an impinging probe particle (neutron) may be assumed to be well defined [3, 2]. Introducing the momentum transfer $\mathbf{q} = \mathbf{k}_0 - \mathbf{k}_1$ from the probe particle to the scattering system, the Fourier transform of the particle density reads

$$n(\mathbf{r}, t) = \frac{1}{(2\pi)^3} \int d\mathbf{q} n(\mathbf{q}, t) \exp\{i \mathbf{q} \cdot \mathbf{r}\} \quad (9)$$

where, in the case of neutron scattering, cf. Eq.(2),

$$n(\mathbf{q}, t) = \sum_j \exp\{-i \mathbf{q} \cdot \mathbf{R}_j(t)\}. \quad (10)$$

Since $n(\mathbf{r}, t)$ is Hermitian it holds $n^\dagger(\mathbf{q}, t) = n(-\mathbf{q}, t)$ and one obtains from Eq. (6)

$$W(t) = \lambda^2 \int_0^t dt' \int_0^t dt'' \sum_f \langle \psi_f | n(\mathbf{q}, t') \rho n(-\mathbf{q}, t'') | \psi_f \rangle, \quad (11)$$

At this stage one traditionally assumes that the sum over ψ_f runs over all possible eigenstates of H_0 which constitute a complete set, i.e. $\sum_f |\psi_f\rangle \langle \psi_f| = 1$; see [3, 2]. Hence

$$\sum_f \langle \psi_f | n(\mathbf{q}, t') \rho n(-\mathbf{q}, t'') | \psi_f \rangle = Tr [n(\mathbf{q}, t') \rho n(-\mathbf{q}, t'')] \quad (12)$$

where $Tr[\dots]$ denotes the trace operation. As done in standard theory [3, 2], in Eq. (12) one first sums over all final states, keeping the initial state ψ_i fixed, and then averages

over all ψ_i (see e.g. [3], p. 19). The right-hand-side of Eq. (12) contains the density operator ρ of the system before collision, Eq. (8), which is a well known result.

By introducing a measurement time (the so-called scattering time) τ_{sc} , that is the duration of the scattering process, we find

$$\begin{aligned} W(\tau_{sc}) &= \lambda^2 \int_0^{\tau_{sc}} dt' \int_0^{\tau_{sc}} dt'' Tr[n(\mathbf{q}, t') \rho n(-\mathbf{q}, t'')] \\ &= \lambda^2 \tau_{sc} \int_0^{\tau_{sc}} d\tau Tr[n(\mathbf{q}, t') \rho n(-\mathbf{q}, t' + \tau)], \end{aligned} \quad (13)$$

where the stationary property of the correlation function has been used [3]. Now one can introduce the transition rate, say \dot{W} , which is defined as

$$\begin{aligned} \dot{W} \equiv \frac{W(\tau_{sc})}{\tau_{sc}} &= \lambda^2 \int_0^{\tau_{sc}} d\tau Tr(n(\mathbf{k}, t') \rho n(-\mathbf{k}, t' + \tau)) \\ &\equiv \lambda^2 \int_0^{\tau_{sc}} d\tau C(\mathbf{q}, \tau). \end{aligned} \quad (14)$$

Here the correlation function

$$C(\mathbf{q}, t) = Tr[n(\mathbf{q}, 0) \rho n(-\mathbf{q}, t)] \quad (15)$$

is introduced, which is analogous to the so-called intermediate function of neutron scattering theory [3].

3 Irreversible dynamics

We now introduce a set of preferred coordinates $\{|\xi\rangle\}$, cf. [9, 10]. These are the relevant degrees of freedom coupled to the neutron probe. The density matrix needed in (13) is then the *reduced* one in the space spanned by these states, and it is obtained by tracing out the (huge number of the) remaining degrees of freedom belonging to the "environment" of the microscopic scattering system (e.g. a proton and its adjacent particles). To simplify notations, we denote this reduced density matrix by ρ too.

In the *subspace* spanned by the preferred coordinates (also denoted 'pointer basis'), we assume the relevant density matrix to obey a Lindblad-type equation of the form [7, 8]

$$\partial_t \rho = -i[H, \rho] + \mathcal{R}\rho \equiv \mathcal{L}\rho \quad (16)$$

with the formal solution

$$\rho(t) = e^{\mathcal{L}t} \rho(0). \quad (17)$$

Let us look at a time-dependent expectation value

$$\langle A(t) \rangle \equiv Tr(\rho(t)A) = Tr(e^{\mathcal{L}t} \rho(0)A) = Tr\left(\rho(0)e^{\mathcal{L}^\dagger t} A\right), \quad (18)$$

where we define \mathcal{L}^\dagger by setting

$$Tr((\mathcal{L}X)Y) = Tr(X(\mathcal{L}^\dagger Y)). \quad (19)$$

Thus we obtain a Lindblad time evolution for the operators too by writing

$$\partial_t A(t) = \mathcal{L}^\dagger A(t). \quad (20)$$

This form was actually the original Lindblad result. Note that this works as long as \mathcal{L} does not depend on time. For time-dependent generators of the evolution, a somewhat more elaborate scheme is needed.

Now we find that we may use this formalism to calculate correlation functions like the one in (14). We write

$$\langle A(t)B \rangle = Tr \left[\rho(0) \left(e^{\mathcal{L}^\dagger t} A \right) B \right] = Tr [A e^{\mathcal{L}^\dagger t} (B \rho(0))] \equiv Tr (A \rho_B(t)), \quad (21)$$

where $\rho_B(t)$, as defined in Eqs. (21), obeys the equation

$$\partial_t \rho_B(t) = \mathcal{L} \rho_B(t) \quad (22)$$

and the initial condition

$$\rho_B(0) = B \rho(0). \quad (23)$$

Thus, except for the initial condition, we have to solve the same equation of motion as for the density matrix (16).

4 Application to scattering

We here assume a simple Lindblad-type ansatz for the master equation in the relevant subspace. We set

$$\partial_t \rho = -i [H, \rho] - K [X, [X, \rho]] = \mathcal{L} \rho, \quad (24)$$

where $K > 0$ and H is the reduced (or relevant Hamiltonian) of a microscopic or mesoscopic scattering system and the double commutator term describes decoherence. For simplicity of the further calculations, we here assume that

$$\begin{aligned} H |\xi\rangle &= \mathcal{E}_\xi |\xi\rangle \\ X |\xi\rangle &= \xi |\xi\rangle. \end{aligned} \quad (25)$$

With Eq. (15) we have

$$\begin{aligned} C(\mathbf{q}, \tau) &= Tr[n(\mathbf{q}, 0) \rho n(-\mathbf{q}, \tau)] = Tr \left(n(\mathbf{q}, 0) \rho e^{\mathcal{L}^\dagger t} n(-\mathbf{q}, 0) \right) \\ &= Tr \left(n(-\mathbf{q}, 0) e^{\mathcal{L}^\dagger t} (n(\mathbf{q}, 0) \rho) \right). \end{aligned} \quad (26)$$

This is equivalent with the expression

$$C(\mathbf{q}, \tau) = \sum_{\xi, \xi'} \langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle \langle \xi' | \rho_n(t) | \xi \rangle. \quad (27)$$

With the equation (24), one easily finds the well known solution

$$\begin{aligned} \langle \xi' | \rho_n(t) | \xi \rangle &= \exp[-i(\mathcal{E}_{\xi'} - \mathcal{E}_\xi)t] \exp[-K(\xi' - \xi)^2 t] \\ &\times \langle \xi' | n(\mathbf{q}, 0) \rho(0) | \xi \rangle. \end{aligned} \quad (28)$$

Inserting this into the expression (14) for the transition rate we find

$$\begin{aligned}\dot{W} = \lambda^2 \int_0^{\tau_{sc}} d\tau \sum_{\xi, \xi'} & \exp[-i(\mathcal{E}_{\xi'} - \mathcal{E}_\xi)t] \exp[-K(\xi' - \xi)^2 t] \\ & \times \langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle \langle \xi' | n(\mathbf{q}, 0) \rho(0) | \xi \rangle.\end{aligned}\quad (29)$$

5 Decoherence and decrease of transition rate

Obviously, the decoherence-free limit of this result, i.e. with $K = 0$, corresponds to the conventional result of scattering theory.

The oscillating factors $\exp[-i(\mathcal{E}_{\xi'} - \mathcal{E}_\xi)t]$ are characteristic for the 'unitary-type' dynamics caused by the commutator part $-i[H, \rho]$ of the master equation (24) for the reduced (or: relevant) density matrix ρ . These factors have the absolute value 1 and do not affect the numerical value of the transition rate.

On the other hand, the restrictive factors $\exp(-K(\xi' - \xi)^2 t) \leq 1$, which are due to the decoherence, can be seen to cause a decrease of the transition rate and thus of the associated cross-section. This can be illustrated in physical terms as follows:

Let us first assume that the reduced density operator $\rho(0)$ can be chosen to be *diagonal* in the preferred ξ -representation (which corresponds to the usual random phase approximation at $t = 0$). Then each term of Eq. (29) is of the form

$$\begin{aligned}\langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle \langle \xi' | n(\mathbf{q}, 0) \rho(0) | \xi \rangle = \\ \langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle \langle \xi' | n(\mathbf{q}, 0) | \xi \rangle \langle \xi | \rho(0) | \xi \rangle = \\ |\langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle|^2 \langle \xi | \rho(0) | \xi \rangle \geq 0.\end{aligned}\quad (30)$$

The last inequality is valid because it holds $\langle \xi | \rho(0) | \xi \rangle \geq 0$.

If the assumed diagonal form of $\rho(0)$ would be considered as being 'too strong', one may note the following: The exponentials $\exp(-K(\xi' - \xi)^2 t)$ due to decoherence imply that only terms with $\xi \approx \xi'$ contribute significantly to the transition rate. Thus we may conclude that, by continuity, all associated terms with $\xi \approx \xi'$ in Eq. (29) should be positive, too. The further terms with ξ being much different from ξ' can be positive or negative. But they may be approximately neglected, since they decay very fast and thus contribute less significantly to \dot{W} .

The main conclusion from the preceding considerations is that the time average in Eq. (29) always decreases the numerical value of $\dot{W} \equiv W(\tau_{sc})/\tau_{sc}$, due to the presence of the exponential factors $\exp(-K(\xi' - \xi)^2 t) \leq 1$. In other words, the effect of decoherence during the experimental time window τ_{sc} plays a crucial role in the scattering process and leads to an 'anomalous' decrease of the transition rate and the associated scattering intensity. This result is in line with that of Ref. [11], which investigated the standard expression of the double differential cross-section of neutron scattering theory by ad hoc assuming decoherence of final and initial states of the scatterer.

Very interesting is also the conclusion that, in the limit of very slow decoherence ($K \rightarrow 0$), this 'anomaly' disappears, i.e. the scattering results are expected to agree

with conventional theoretical expectations. This is contrary to the associated prediction of the theoretical model of Ref. [12]

6 Additional remarks

A related effect (i.e., a shortfall of scattering intensity) was observed in recent neutron-proton Compton scattering (NCS) and electron-proton Compton scattering (ECS) experiments in condensed matter [5, 6], in which the experimental scattering time lies in the sub-femtosecond time scale. This coincides with the characteristic time of electronic re-arrangements accompanying the breaking (or formation) of a chemical bond. Note that in these experiments the energy transferred to a proton is large enough to break the bond (C–H and O–H).

Some remarks about the possible selection, definition and/or physical meaning of the preferred coordinates may be appropriate. In the case of conventional NCS theory, for example, one uses momentum eigenstates of the scattering particle (e.g. proton) — as well as for the neutron — as the appropriate basis [13]. In the light of the preceding derivations, however, one may observe the following: Due to the strong (Coulomb) interactions of the scattering proton with its adjacent particles (electrons, and probably also other nuclei), $\{|\xi\rangle\}$ can not be one-body states but they should rather be considered to represent momentum states being strongly "dressed" (and entangled) with degrees of freedom of adjacent particles.

Further work will deal with the more general — and experimentally relevant — case, in which the preferred states $\{|\xi\rangle\}$ are not eigenstates of the "reduced" energy and Lindblad operators, Eqs. (25). In that case, the result of Eq. (29) will become less simple.

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References

- [1] A. Einstein, B. Podolsky and N. Rosen, Phys. Rev. **47**, 777 (1935).
- [2] L. van Hove, Phys. Rev. **95**, 249 (1954).
- [3] G. L. Squires, *Introduction to the Theory of Thermal Neutron Scattering* (Dover, Mineola, 1996).
- [4] E. Weigold and I. E. McCarthy, *Electron Momentum Spectroscopy* (Kluwer Academic/Plenum, New York, 1999).
- [5] C. A. Chatzidimitriou-Dreismann et al., Phys. Rev. Lett. **79**, 2839 (1997); and **91**, 057403 (2003).

- [6] (a) Physics Today, Physics Update, p. 9, September 2003; (b) Scientific American, p. 20, October 2003; (c) The AIP Bulletin of Physics News, Physics New Update No. 648, 31 July 2003.
- [7] G. Lindblad, Comm. Math. Phys. **48**, 119 (1976).
- [8] S. M. Barnett and S. Stenholm, Phys. Rev. A **64**, 033808 (2001).
- [9] (a) H. D. Zeh, Found. Phys. **3**, 109 (1973); (b) O. Kübler and H. D. Zeh, Ann. Phys. (NY) **76**, 405 (1973).
- [10] (a) W. H. Zurek, Phys. Rev. D **24**, 1516 (1981); and **26**, 1862 (1982); (b) W. H. Zurek, Rev. Mod. Phys. **75**, 715 (2003).
- [11] C. A. Chatzidimitriou-Dreismann, preprint (2003); and J. Alloys Compd. **356-357**, 244 (2003)
- [12] E. B. Karlsson and S. W. Lovesey, Phys. Rev. A **61**, 062714 (2000); Phys. Scr. **65**, 112 (2002).
- [13] G. I. Watson, J. Phys. Condens. Matter **8**, 5955 (1996).